

REMARKS

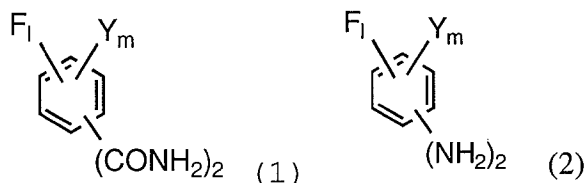
This document is submitted in reply to the Office Action dated June 6, 2008 (“Office Action”).

Applicants respectfully request that the Examiner reconsider this application in view of the following remarks.

I

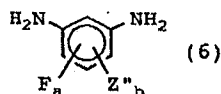
Claims 1-12 are rejected as obvious over Masayoshi et al., European Patent Application No. 1275679 (“Masayoshi”), in view of Andrews et al., Aust. J. Chem., 1971, 412-422 (“Andrews”), and Hazen et al., U.S. Patent No. 5,011,997 (“Hazen”). See the Office Action, page 2, lines 10-16.

Independent claim 1 will be discussed first. This claim covers a method for obtaining a fluorinated phenylenediamine of formula (2) shown below. The fluorinated diamine (2) is obtained via a Hoffman rearrangement by reacting a diamide, represented by formula (1) below, with NaOX, where X is either Br or Cl, in the presence of NaOH. In this method, the molar ratio of NaOH to diamide is in the range of 1.8-6.0.



Masayoshi, the primary reference, teaches compounds of formula 2, but does not teach any method of making them from diamides. Indeed, the Examiner concedes that “Masayoshi does not teach a method for the production of a fluorinated aromatic diamine from corresponding diamide.”¹ See the Office Action, page 3, last sentence. In other

¹ According to the Examiner, Masayoshi discloses “a fluorinated phenylenediamine of ... formula (6) where Z” denotes a chlorine, bromine or iodine ... *a* denotes fluorine atoms bonded to a benzene ring, representing an integer of 0-4 ... and *b* denotes the number of ‘Z’” bonded to a benzene ring, representing 0-4 ... the total of *a* and *b* ought to be invariably 4.” See the Office Action, page 2, line 17 through page 3, line 7, *a* and *b* have been italicized.



words, Masayoshi does not teach obtaining a diamine from its corresponding diamide, as required by claim 1, let alone using a molar ratio of NaOH to diamide in the range of 1.8-6.0, as also required by claim 1.

Turning to Andrews, a secondary reference, the Examiner states that it discloses preparing an isophthalic-based diamine from a corresponding diamide with the Hoffmann rearrangement. The Examiner also states that Andrews teaches that the NaOH/diamide ratio is 2.25, which is within the range of claim 1. See the Office Action, page 4, lines 11-13.

Applicants would like to point out that Andrews discloses two methods which use the Hoffmann rearrangement for preparing an isophthalic-based diamine from its corresponding diamide. In the first method, the NaOH/diamide ratio was 2.25. This method resulted in a very low diamine yield of only 19%. See page 416, lines 23-38.

In the second method, characterized in Andrews as “a more efficient preparation” (see page 416, line 38), a NaOH/diamide ratio of about 1.56 was used, which resulted in a significantly increased diamine yield of 74%. See page 416, lines 39-47.

The NaOH/diamide ratio in the second method was calculated to be about 1.56 as follows (based on three facts/assumptions, i.e., [i] 3.38 g of 2-nitroisophthalic acid diamide and 100 mL of 1% aqueous NaOH solution was used,² [ii] the molecular weights of 2-nitroisophthalic acid diamide and NaOH are 211 g/mole and 40 g/mole, respectively, and [iii] the density of the 1% aqueous NaOH solution is identical to that of water, i.e., 1 g/ml):

$$\begin{aligned}\text{Moles of NaOH} &= (100 \text{ ml} \times 1\% \times 1 \text{ g/ml}) / 40 \text{ g/mole} = \mathbf{0.025}; \\ \text{Moles of diamide} &= 3.38 \text{ g diamide} / 211 \text{ g/mole} = \mathbf{0.016}; \\ \text{NaOH/diamide ratio} &= \mathbf{0.025/0.016} = 1.56.\end{aligned}$$

In sum, Andrews teaches that the first method having a NaOH/diamide ratio of 2.25 resulted in a 19% diamine yield, while the second having a NaOH/diamide ratio of about 1.56, resulted in a significantly improved yield of 74%. Given this teaching, a skilled artisan would have been motivated by the vastly improved yield to lower the

² For the amount of diamide and the volume of 1% aqueous NaOH solution used in Andrew's second method, see page 416, lines 39 and 41, respectively.

NaOH/diamide ratio from 2.25 to 1.56, not increase it to a range of 1.8-6.0 as required by claim 1. Clearly, Andrew teaches away from using a NaOH/diamide ratio in the range of 1.8 to 6.0.

Hazen, the other secondary reference, discloses a method for producing a fluorinated aromatic diamine from its corresponding diamide. See the Abstract. As correctly pointed out by the Examiner, the Hoffmann rearrangement is “the same method used by both Hazen and the Application examined.” See the Office Action, page 4, lines 6-8. As also correctly pointed out by the Examiner, in the Hazen’s method, a molar ratio of the NaOH to the diamide is higher than 6.0. See the Office Action, page 4, lines 5-6. Clearly, Hazen does not teach or suggest a yield/purity trend as a function of NaOH/diamide ratio, let alone a NaOH/diamide ratio in the range of 1.8 to 6.0, as required by claim 1.

To conclude, Masayoshi does not teach using a NaOH/diamide ratio of 1.8 to 6.0 in producing a diamine from a diamide, as required in the method claim 1, and neither Andrews nor Hazen cures this deficiency. Thus, the combination of these three references does not render claim 1 obvious. Nor does their combination render obvious claims 2-4 and 7-10, all of which depend, directly or indirectly, from claim 1. Clearly

Applicants now turn to the rejection of independent claims 5, 6, 11, and 12. Claims 5 and 11 each cover a method for obtaining a polyamic acid while claims 6 and 12 each cover a method for obtaining a polyimide. The first step in each method is identical to that of claim 1, which is discussed above. Thus, for the same reasons and facts set forth above, claims 5, 6, 11, and 12 are also not rendered obvious by the combination of Masayoshi, Andrews, and Hazen.

II

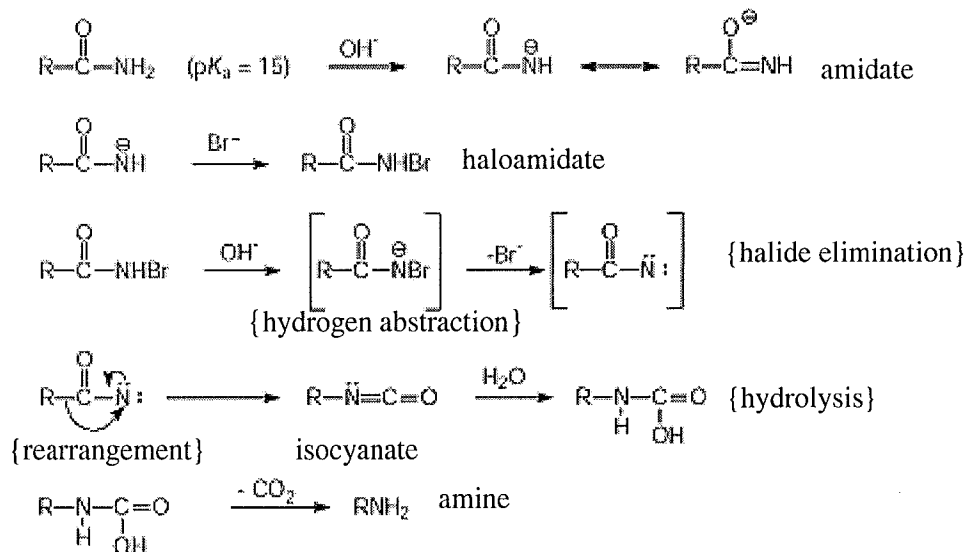
As will be discussed below, even if the Examiner had established a prima facie case of obviousness (which Applicants do not concede) in view of the combination of Masayoshi, Andrews, and Hazen, it could be successfully rebutted by a showing of unexpected results.

Applicants argued in their last response dated January 14, 2008 ("Response") that their method, which used a NaOX/diamide ratio in the range of 3.0-6.0, compared with Hazen's which used a ratio below 3.0, resulted in a more pure product with higher yields and that these unexpected results could rebut the Examiner's prima facie case of obviousness.

After reviewing Applicants' arguments, the Examiner asserts that Applicants' and Hazen's methods "are not comparable, since Hazen teaches a process for bis(4-aminophenyl) hexafluoropropane, whereas in Example 2, the Applicant produces tetrafluoroisophthalamide." See the Office Action, page 7, lines 16-20. It is clearly the Examiner's position that, as the diamides used in Applicants' and Hazen's methods are different, the corresponding products yield/purity trends cannot be relied on to show unexpected results. Applicants respectfully disagree.

As will be discussed below, the yield/purity trends, based on both NaOX/diamide and NaOH/diamide ratios, for diamines produced from different diamides are comparable.

As shown in the scheme below, in the Hoffmann rearrangement, NaOH reacts with the nitrogen atom of an $-\text{CONH}_2$ group to form an amidate, the amidate subsequently reacts with a halogen to form a haloamidate, hydrogen on the haloamidate is then abstracted by NaOH, resulting in halide elimination, and finally a molecular rearrangement takes place to yield an isocyanate, which is then hydrolyzed and loses $-\text{CO}_2$ to produce an amine.



As can be seen from the above scheme, NaOH and NaOX always react with an amide to form an amine. In other words, both NaOH and NaOX will react with any amide in the same way regardless of the amide's specific chemical structure. As the structure of the amide does not change how both NaOH and NaOX will react with it, a yield/purity trend based on NaOX/amide and NaOH/amide ratios for one amide is comparable to that of a different amide. Thus, contrary to the Examiner's assertion, Applicants' and Hazen's methods are comparable and that a yield/purity trend based on NaOX/diamide ratios can be relied on to show unexpected results over those in Hazen. See the Response page 12, line 21 through page 13, line 11.

As discussed below, Applicants' use of a NaOH/diamide ratio in the range of 1.8 to 6.0 also leads to unexpected results. In Applicants' method, the reactions in the Hoffmann rearrangement can be seen to take place in two stages. The first stage includes all of the reactions leading to the rearrangement and formation of isocyanate. During this first stage, the NaOH/diamide ratio is kept in the above-mentioned range. During the second stage, i.e., the hydrolysis process that converts the isocyanate to an amine, an additional amount of NaOH is added. In contrast, the Hazen method does not differentiate between the rearrangement and the subsequent hydrolysis process, as seen from their initial use of an excess quantity of NaOH and lack of a later NaOH addition.

Applicants showed in Comparative Example 1 the effects of an excess quantity of NaOH initially used on the reaction yield. See the Specification, page 28, line 28 through page 29, line 23. In this example, during the Hoffmann rearrangement the NaOH/diamide ratio was 7.02. The use of an excess amount of NaOH during this first stage is similar to Hazen's method. This approach resulted in a significantly reduced diamine yield of 16.4% when compared with the yields of 63-80.7% obtained when the NaOH/diamide ratio was in the range of 1.8 to 6.0. See the Specification Examples 1-3, pages 23-26. Furthermore, the diamine purity shown in Examples 1-3 was in excess of 99.8%, vastly higher than that described for the diamine product of the Hazen method, and as a result, there was no need to conduct a hydrogenation reaction to remove any by-products as taught in Hazen.

For the reasons and facts discussed above, Applicants submit that not only are their and Hazen's yield/purity trends based on NaOX/diamide and NaOH/diamide ratios are comparable, but Applicants' use of NaOX/diamide ratios in the range of 3.0-6.0 and of NaOH/diamide ratios in the range of 1.8-6.0 results in significant improvements in diamine yields and purities.

III

To complete the record, Applicants would like to address a statement made by the Examiner.

According to the Examiner, Hazen teaches that any amount of NaOX may be used in the Hoffmann rearrangement "from theoretical to large excess in order to achieve a balance of purity of product, its yield and severity of recitation conditions." See the Office action, page 5, lines 13-17.

Admittedly, Hazen does suggest the use of an immensely broad range of NaOX/diamide ratios in this reaction. However, Applicants would like to point out that Hazen does not specifically disclose the NaOX/diamide ratio of about 3.0-6.0 recited in claim 1. More importantly, as discussed above, the patentability of claim 1 resides at least in part in "a NaOH/diamide ratio within the range of 1.8 to 6.0," not "a NaOX/diamide ratio in the range of 3.0 to 6.0."

CONCLUSION

It is believed that all of the pending claims have been addressed. However, the absence of a reply to a specific rejection, issue or comment does not signify agreement with or concession of that rejection, issue or comment.

In addition, because the arguments made above may not be exhaustive, there may be reasons for patentability of any or all pending claims (or other claims) that have not been expressed.

Finally, nothing in this paper should be construed as an intent to concede any issue with regard to any claim, except as specifically stated in this paper, and the amendment of any claim does not necessarily signify concession of unpatentability of the claim prior to its amendment.

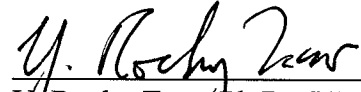
Applicant(s) : Shinji Nishimae et al.
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Attorney Docket No.: 60004-109US1

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Respectfully submitted,

Date: 8-5-08


Y. Rocky Tsao, Ph.D., J.D.
Attorney for Applicants
Reg. No. 34,053

Occhiuti Rohlicek & Tsao LLP
10 Fawcett Street
Cambridge, MA 02138
Telephone: (617) 500-2509
Facsimile: (617) 500-2499
46148.doc